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Radiative Transfer Model for the Reflectance of Packed Particulate Media

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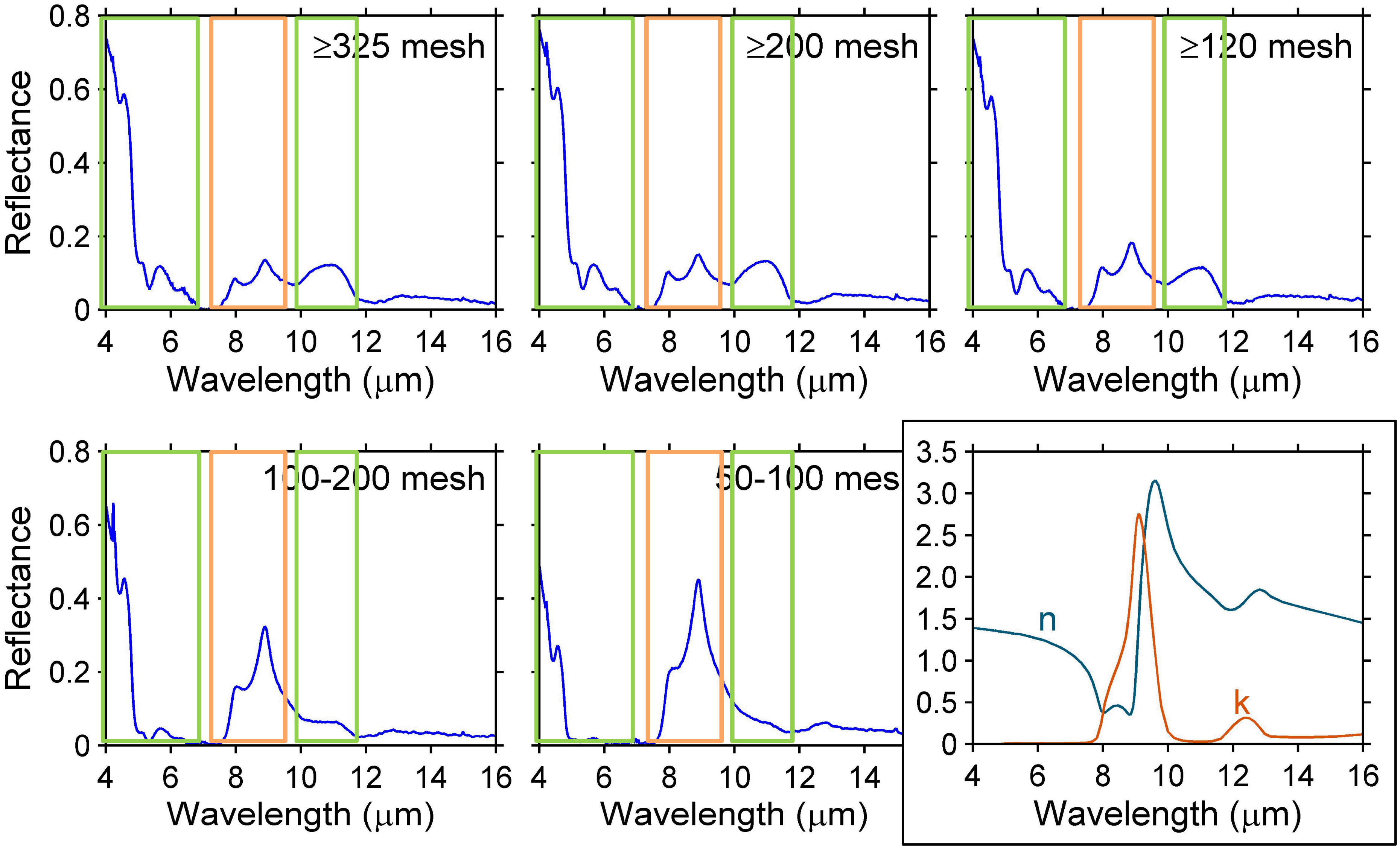
March 21st, 2017

Silica Powders: An Infrared Example of the Morphological Dependence



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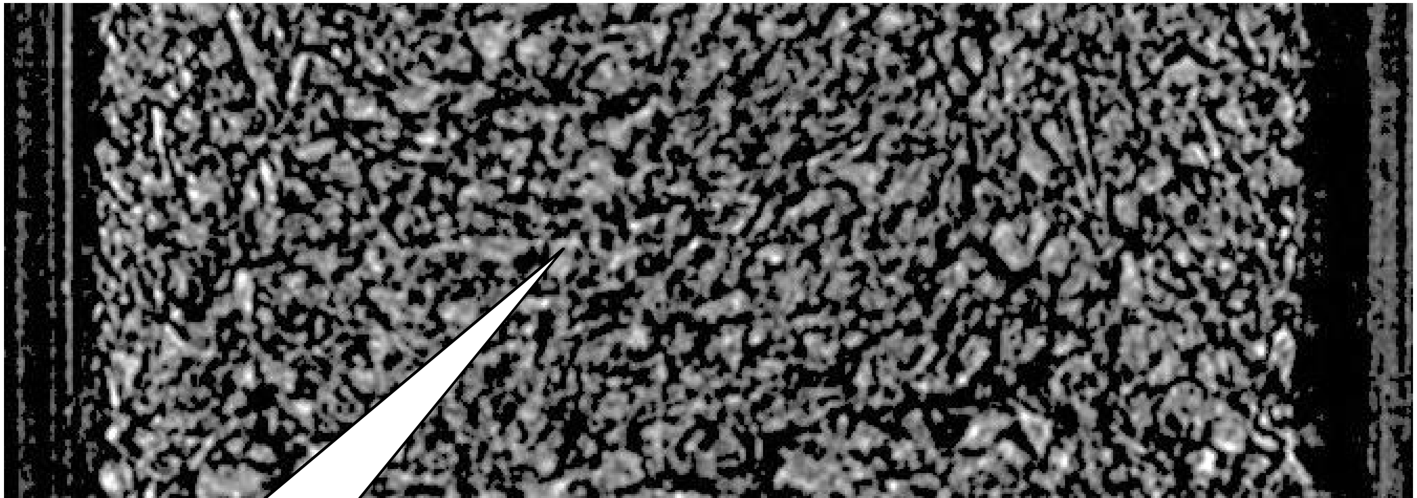
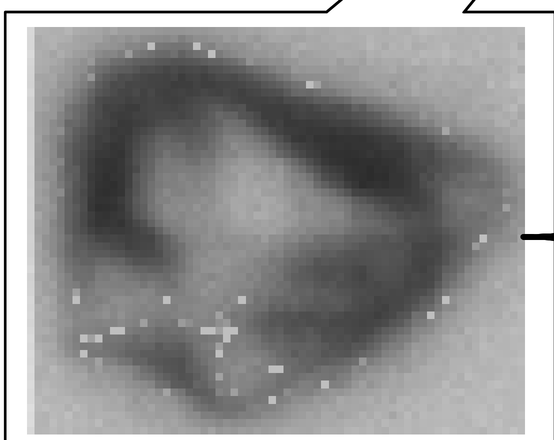
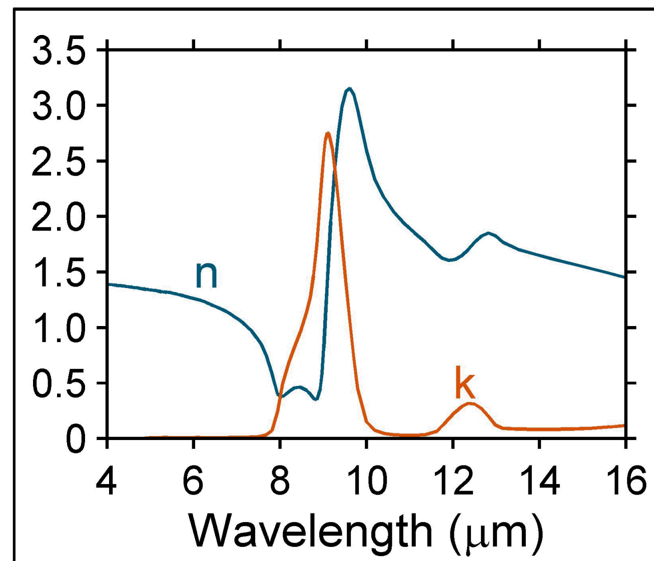




$$\nabla \cdot \mathbf{D}(\mathbf{r}) = \rho(\mathbf{r})$$
$$\nabla \times \mathbf{E}(\mathbf{r}) = i\omega\mu\mathbf{H}(\mathbf{r})$$
$$\nabla \cdot [\mu\mathbf{H}(\mathbf{r})] = 0$$
$$\nabla \times \mathbf{H}(\mathbf{r}) = \mathbf{J}(\mathbf{r}) - i\omega\mathbf{D}(\mathbf{r})$$
$$= -i\omega\varepsilon\mathbf{E}(\mathbf{r})$$

$$n + ik = c(\varepsilon\mu)^{1/2}$$



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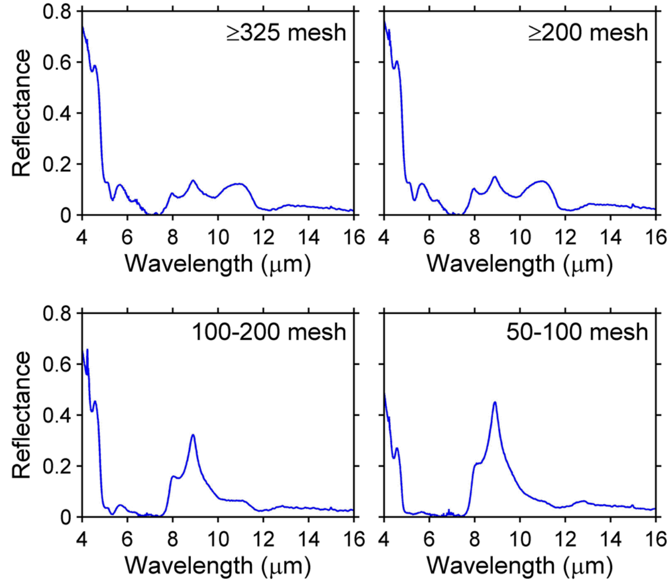
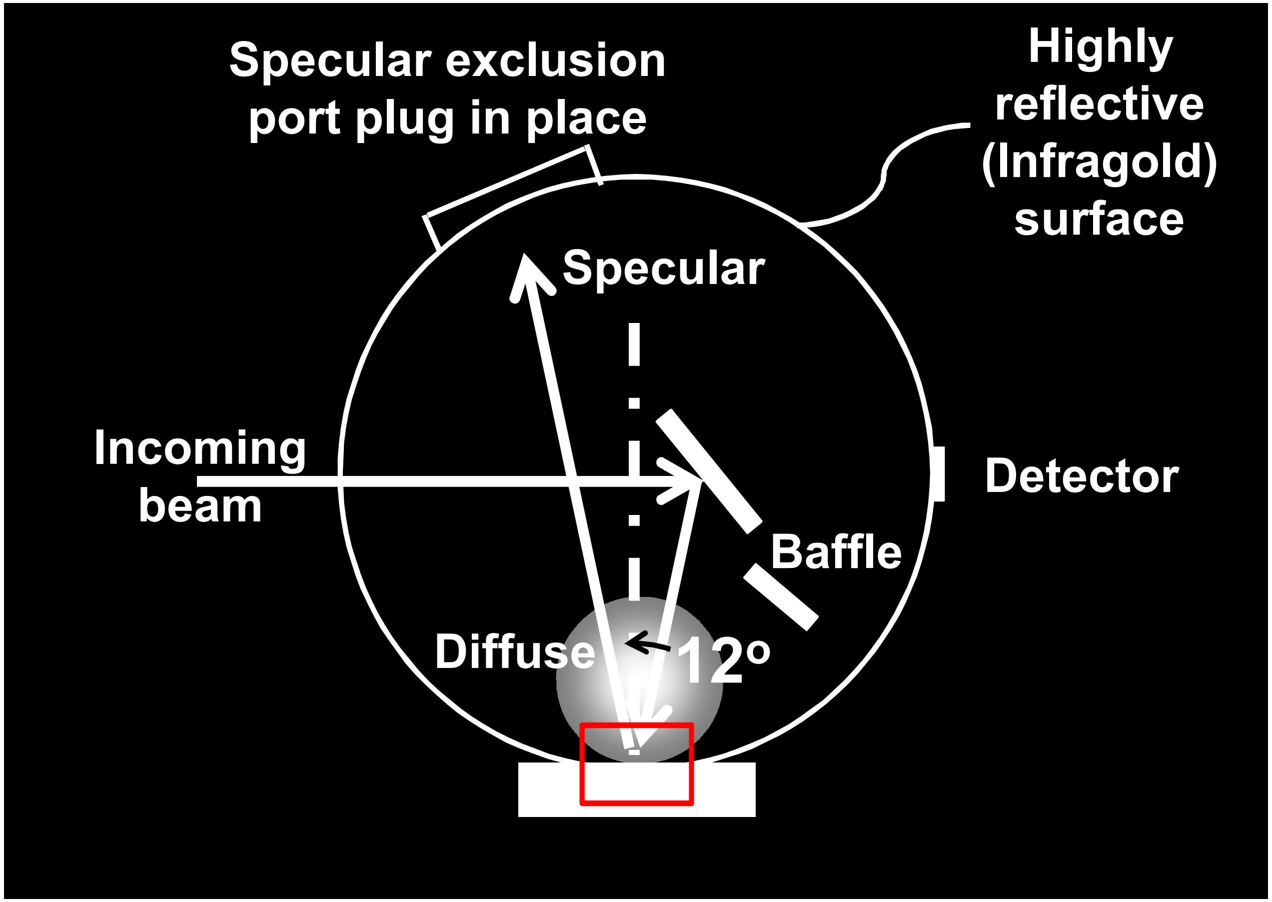
Our Goal, Method, and Focus

- **Goal:** Generate morphologically dependent spectra via physics-based modeling
- **Method:** Develop radiative transfer (RT) model w/parametric inputs to be varied in optimizing agreement w/measurements
  - Incorporate fundamental physical properties (*n*,*k*) and account for morphological characteristics with sufficient physical rigor
  - Optimized, the model should demonstrate agreement with reflectance spectra…
  - …while the extracted parameters should agree with independent measurements
- **Focus:** Match model to reflectance spectra of silica powders
- ***Call your attention to model approximations***



Mesh	Size (μm)
325	44
200	74
120	125
100	149
50	297

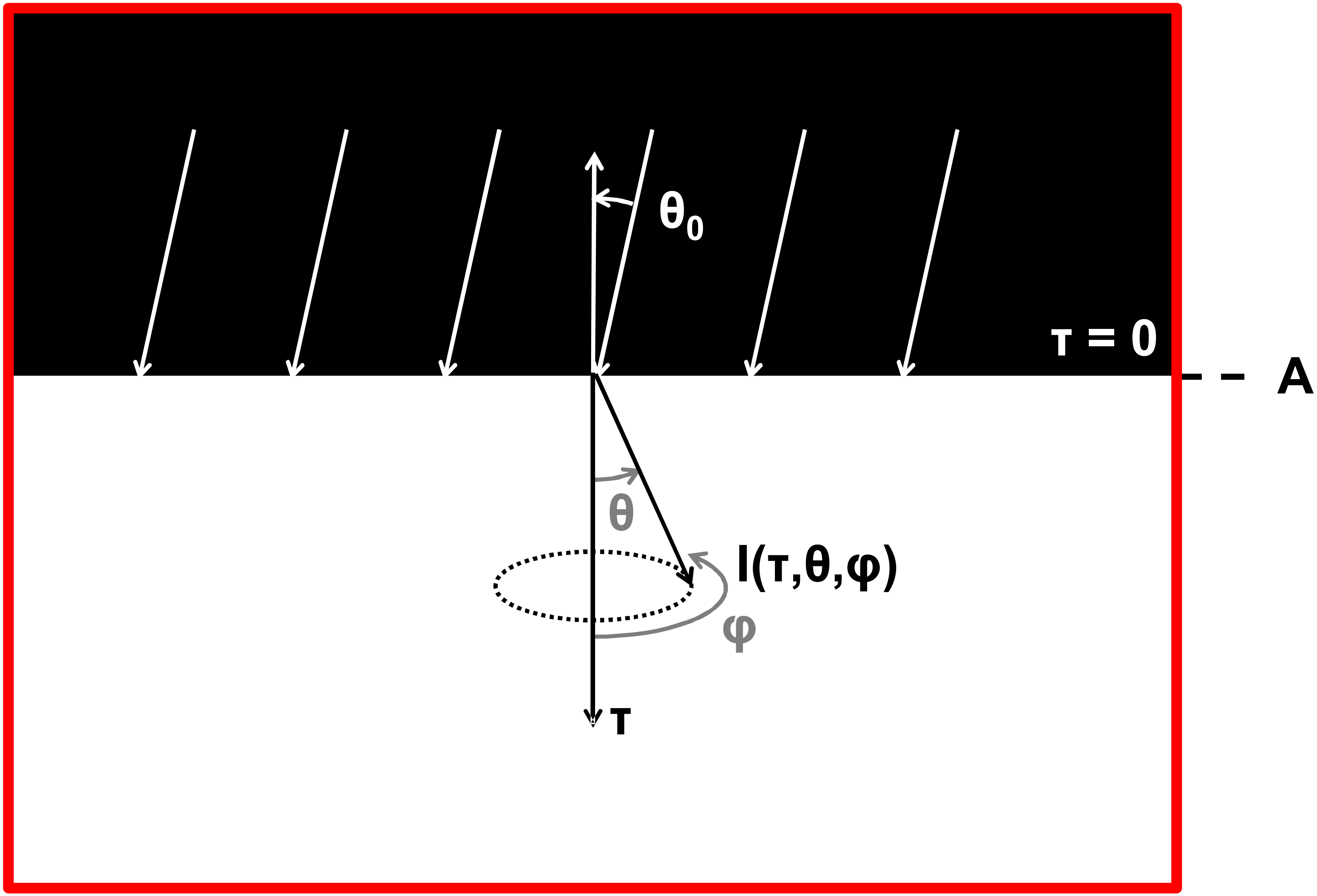
Plane Albedo (Directional, Hemispherical Reflectance) Measurement



Measuring plane albedo with 12° incidence angle

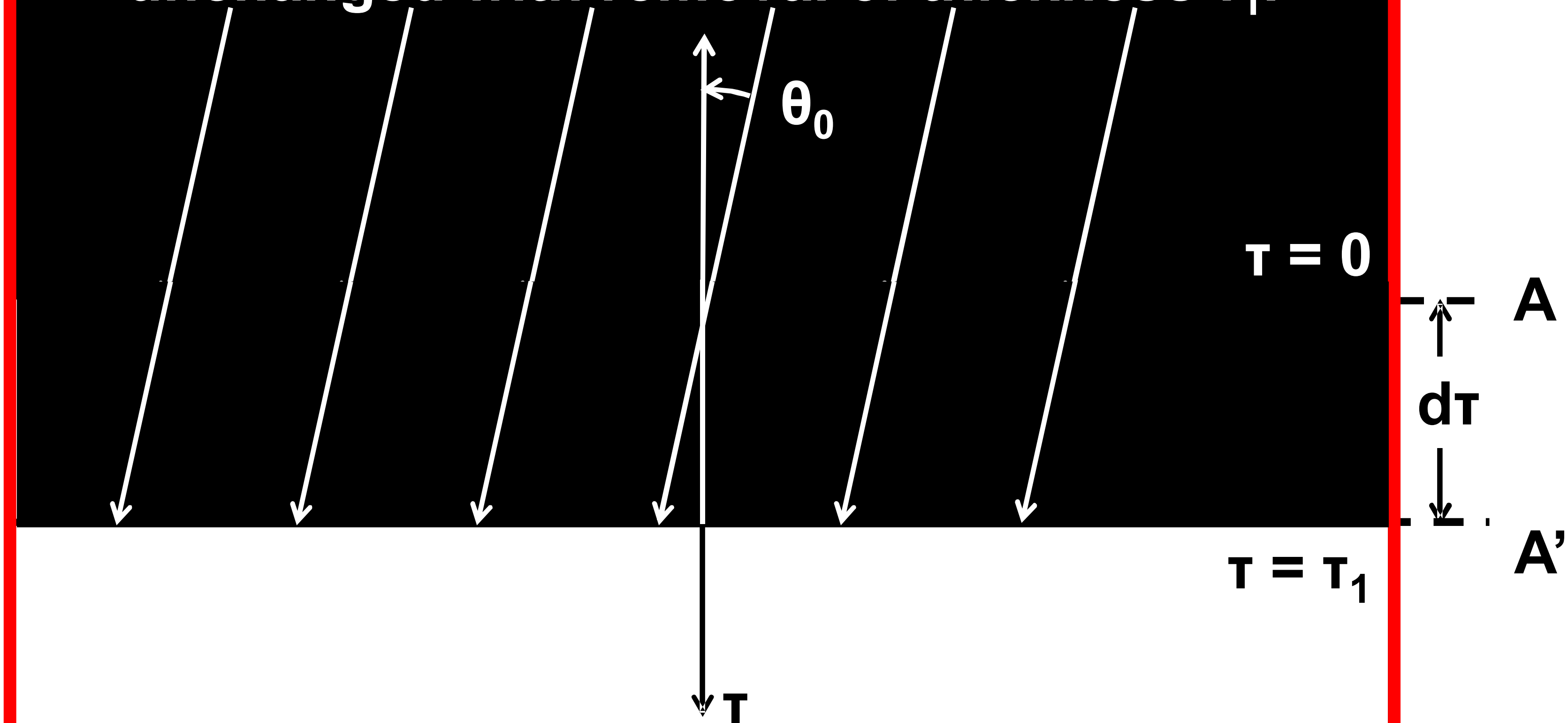
Invariant Imbedding Solution

Invariant Imbedding Solution



Invariant Imbedding Solution

Invariance relationship: Reflectance is unchanged with removal of thickness τ_1.



Solve for reflectance *R* as a Fourier series in azimuth

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Ambartsumian Nonlinear Integral Equation Solves for Fourier Coefficients of Reflectance R^m

E. G. Yanovitskij, *Light scattering in Inhomogeneous Atmospheres*, Trans. by S. Ginsheimer and O. Yanovitskij, Springer (1997).

$$
\begin{aligned}
(\mu+\mu_0)R^m(\mu,\mu_0) = {} & \frac{\varpi}{4}P^m(-\mu,\mu_0) \\
& + \frac{\varpi}{2}\mu_0\int_0^1 P^m(\mu,\mu')\,R^m(\mu',\mu_0)\,d\mu' \\
& + \frac{\varpi}{2}\mu\int_0^1 R^m(\mu,\mu')\,P^m(\mu',\mu_0)\,d\mu' \\
& + \varpi\mu\mu_0\int_0^1\int_0^1 R^m(\mu,\mu')\,P^m(-\mu',\mu'') \\
& \cdot R^m(\mu'',\mu_0)\,d\mu'\,d\mu''
\end{aligned}
$$

$\mu = \left|\cos\begin{pmatrix}\text{observation}\\ \text{angle}\end{pmatrix}\right|$, $\mu_0 = \left|\cos\begin{pmatrix}\text{incident}\\ \text{angle}\end{pmatrix}\right|$

Fourier coefficients of particle scattering function — **Single scattering albedo**

$$P^m,\varpi \;\longrightarrow\; R^m$$

Solution Provided by Michael Mishchenko (NASA GISS)

PERGAMON

Journal of Quantitative Spectroscopy & Radiative Transfer 63 (1999) 409–432

Journal of Quantitative Spectroscopy & Radiative Transfer

www.elsevier.com/locate/jqsrt

Bidirectional reflectance of flat, optically thick particulate layers: an efficient radiative transfer solution and applications to snow and soil surfaces

Michael I. Mishchenko[a,*], Janna M. Dlugach[b], Edgard G. Yanovitskij[b], Nadia T. Zakharova[c]

[a]NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA
[b]Laboratory for Radiative Transfer Theory, The Main Astronomical Observatory, Ukrainian National Academy of Sciences, Golosiiv, 252650 Kyiv-22, Ukraine
[c]Science Systems and Applications, Incorporated, 2880 Broadway, New York, NY 10025, USA

Abstract

We describe a simple and highly efficient and accurate radiative transfer technique for computing bidirectional reflectance of a macroscopically flat scattering layer composed of nonabsorbing or weakly absorbing, arbitrarily shaped, randomly oriented and randomly distributed particles. The layer is assumed to be homogeneous and optically semi-infinite, and the bidirectional reflection function (BRF) is found by a simple iterative solution of the Ambartsumian's nonlinear integral equation. As an exact solution of the radiative transfer equation, the reflection function thus obtained fully obeys the fundamental physical laws of energy conservation and reciprocity. Since this technique bypasses the computation of the internal radiation field, it is by far the fastest numerical approach available and can be used as an ideal input for Monte Carlo procedures calculating BRFs of scattering layers with macroscopically rough surfaces. Although the effects of packing density and coherent backscattering are currently neglected, they can also be incorporated. The FORTRAN implementation of the technique is available on the World Wide Web at http://www.giss.nasa.gov/ ~ crmim/brf.html and can be applied to a wide range of remote sensing, engineering, and biophysical problems. We also examine the potential effect of ice crystal shape on the bidirectional reflectance of flat snow surfaces and the applicability of the Henyey–Greenstein phase function and the δ-Eddington approximation in calculations for soil surfaces. © 1999 Elsevier Science Ltd. All rights reserved.

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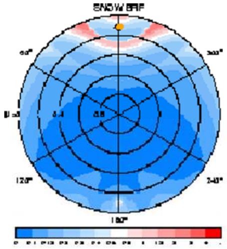
Electromagnetic Scattering by Particles and Surfaces

FORTRAN Codes for the Computation of the Bidirectional Reflection Function for Flat Particulate Layers and Rough Surfaces

By Michael I. Mishchenko and Nadia T. Zakharova



This webpage provides access to two collections of FORTRAN codes.

The first one can be used to compute the (scalar) bidirectional reflectance of a semi-infinite homogeneous slab composed of arbitrarily shaped, randomly oriented particles based on a rigorous numerical solution of the radiative transfer equation.

The second one can be used to compute the Stokes reflection matrix of a rough interface separating two homogeneous half-spaces with different refractive indices (e.g., a rough ocean surface).

Particulate Semi-Infinite Layers
The code brf.f solves the Ambartsumian's nonlinear integral equation for the reflection function using a simple iterative method. Since this technique bypasses the computation of the internal field, it is by far the fastest and most accurate numerical approach available.

The codes are ideally suitable to computing the BRF for flat snow, soil, and powder surfaces and optically thick clouds and may find applications in geophysics, physics, biophysics, and industrial research.

A detailed user manual to the codes has been published: M. I. Mishchenko, J. M. Dlugach, E. G. Yanovitskij, and N. T. Zakharova, Bidirectional reflectance of flat, optically thick particulate layers: An efficient radiative transfer solution and applications to snow and soil surfaces, *J. Quant. Spectrosc. Radiat. Transfer*, **63**, 409-432 (1999). A hardcopy reprint of this paper is available from Michael Mishchenko upon request. Please leave a message at mmishchenko@giss.nasa.gov indicating your name and mailing address.

The users of the codes are encouraged to visit this page on a regular basis for information on latests developments, warnings, and/or errors found. We would highly appreciate informing us of any problems and errors encountered with these codes. Please e-mail your questions and comments to mmishchenko@giss.nasa.gov.

FORTRAN codes
To retrieve a code, click on the code name and use the "Save As..." option from the "File" menu.

- refl.f - This code computes Fourier components of the reflection function
- interp.f - This code computes the bidirectional reflection function for a given set of scattering geometries
- spher.f - This code computes the Legendre expansion coefficients for polydisperse spherical particles using the standard Lorenz-Mie theory

The codes must be run in the following sequence: spher.f -> refl.f -> interp.f.

Note that the Legendre expansion coefficients for polydisperse, randomly oriented nonspherical particles and sphere aggregates can be computed using O T-matrix codes also available on this website. The expansion coefficients for the standard and double-peaked Henyey-Greenstein phase functions are computed using Eqs. (15) and (19) of the manual. Below we also provide the Legendre expansion coefficients for two nonspherical ice particle models described in the manual.

Benchmark results
The following output files were computed by the codes in their current settings and may provide a useful test of the performance of the codes on different computers:

- spher.print
- spher.write
- refl.print
- interp.write

The file refl.write is not given here because of its large size.

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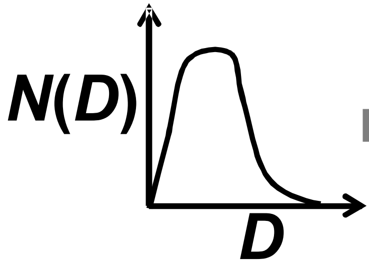
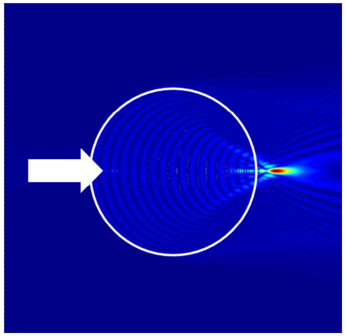
Solution Provided by Michael Mishchenko (NASA GISS)

Step #1: *n*,*k* + 

→ P^m, $\tilde{\omega}$



Step #2: P^m, $\tilde{\omega}$ → R^m

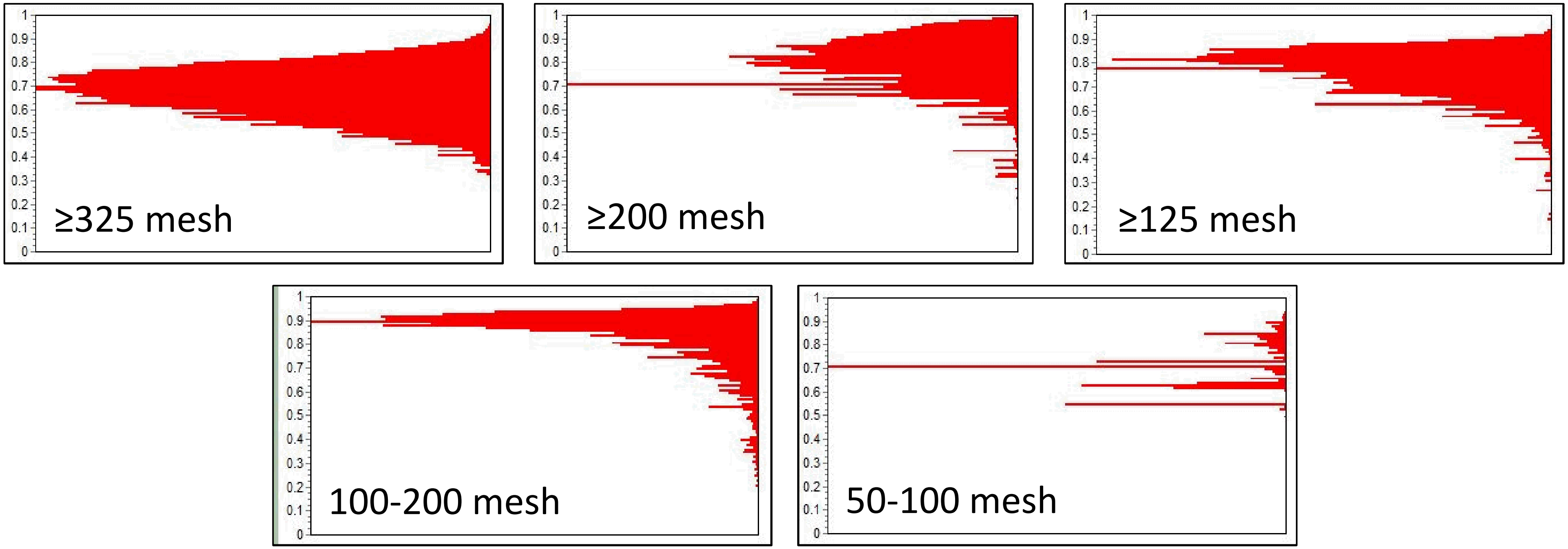
$$(\mu+\mu_0)R^m(\mu,\mu_0)=\frac{\varpi}{4}P^m(-\mu,\mu_0)+\frac{\varpi}{2}\mu_0\int_0^1 P^m(\mu,\mu')\,R^m(\mu',\mu_0)\,d\mu'$$
$$+\frac{\varpi}{2}\mu\int_0^1 R^m(\mu,\mu')\,P^m(\mu',\mu_0)\,d\mu'$$
$$+\varpi\mu\mu_0\int_0^1\int_0^1 R^m(\mu,\mu')\,P^m(-\mu',\mu'')\cdot R^m(\mu'',\mu_0)\,d\mu'\,d\mu''$$

Step #3:

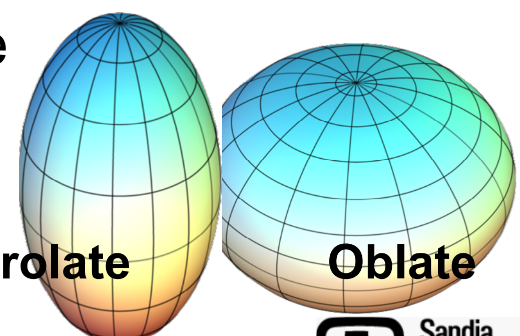
$$R(\mu,\mu_0,\varphi)=R^0(\mu,\mu_0)+2\sum_{m=1}^{m_{\max}}R^m(\mu,\mu_0)\cos(m\varphi)$$

$$\text{Plane albedo}(\mu_0)=2\int_0^1 R^0(\mu,\mu_0)\,\mu\,d\mu$$

Our Canonical Geometry: Spheroids



- This range of shapes and aspect ratios (ARs) is greatly simplified
- Shape = spheroid, AR = one characteristic value
- 2 shape bins: one bin each per prolate, oblate



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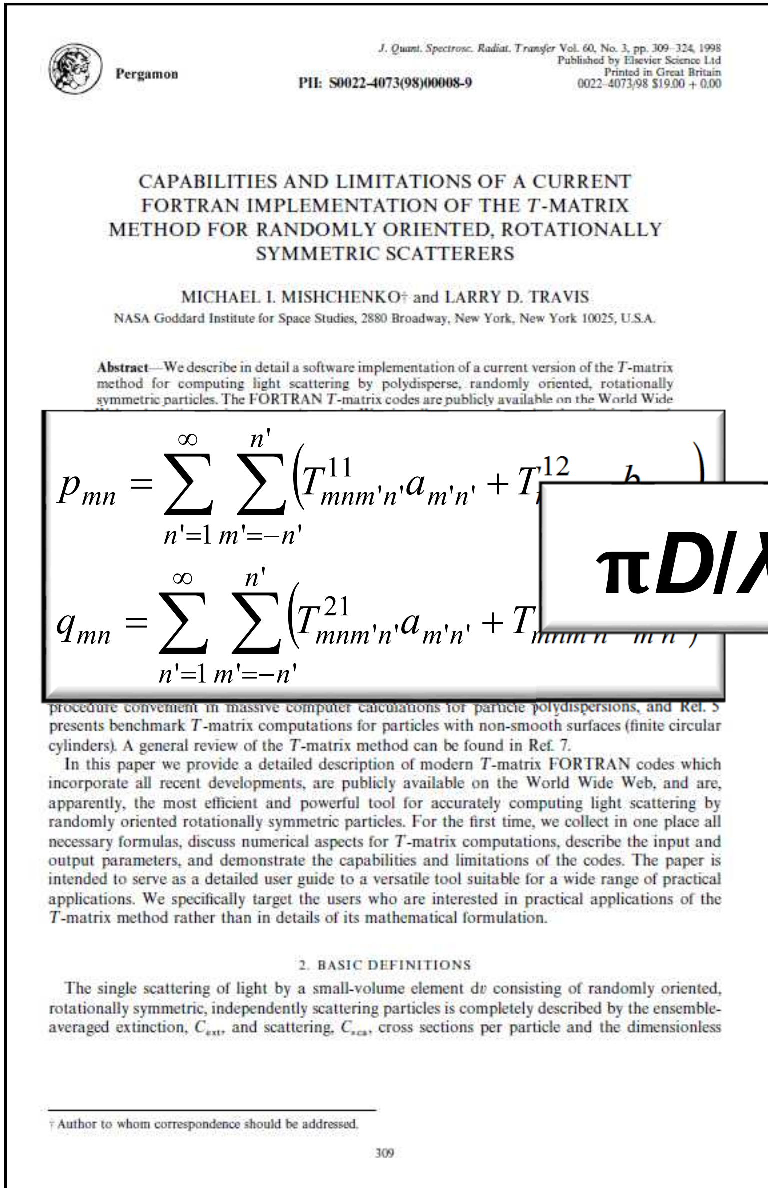
Calculating P^m, $\tilde{\omega}$


Pergamon
J. Quant. Spectrosc. Radiat. Transfer Vol. 60, No. 3, pp. 309–324, 1998
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CAPABILITIES AND LIMITATIONS OF A CURRENT FORTRAN IMPLEMENTATION OF THE *T*-MATRIX METHOD FOR RANDOMLY ORIENTED, ROTATIONALLY SYMMETRIC SCATTERERS

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NASA Goddard Institute for Space Studies, 2880 Broadway, New York, New York 10025, U.S.A.

Abstract—We describe in detail a software implementation of a current version of the *T*-matrix method for computing light scattering by polydisperse, randomly oriented, rotationally symmetric particles. The FORTRAN *T*-matrix codes are publicly available on the World Wide

presents benchmark *T*-matrix computations for particles with non-smooth surfaces (finite circular cylinders). A general review of the *T*-matrix method can be found in Ref. 7.

In this paper we provide a detailed description of modern *T*-matrix FORTRAN codes which incorporate all recent developments, are publicly available on the World Wide Web, and are, apparently, the most efficient and powerful tool for accurately computing light scattering by randomly oriented rotationally symmetric particles. For the first time, we collect in one place all necessary formulas, discuss numerical aspects for *T*-matrix computations, describe the input and output parameters, and demonstrate the capabilities and limitations of the codes. The paper is intended to serve as a detailed user guide to a versatile tool suitable for a wide range of practical applications. We specifically target the users who are interested in practical applications of the *T*-matrix method rather than in details of its mathematical formulation.

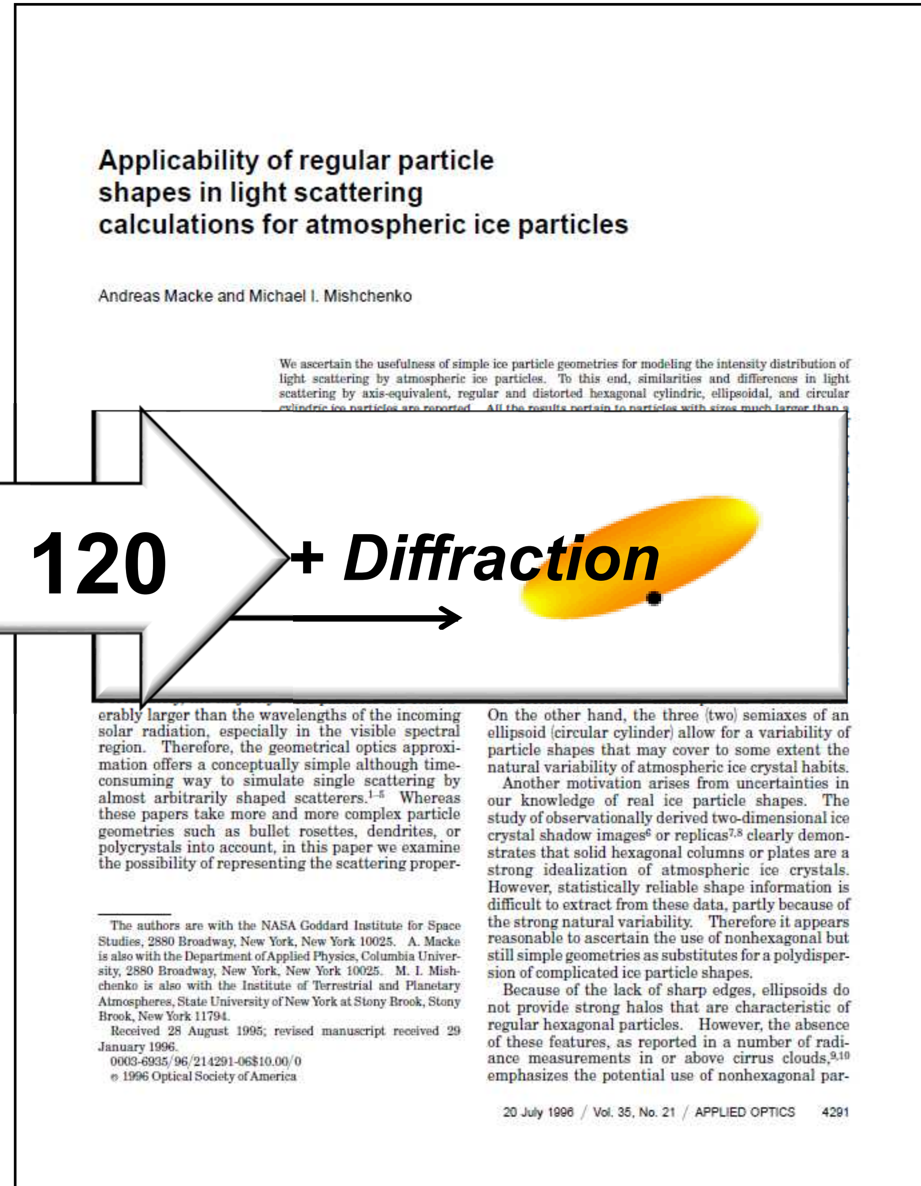
2. BASIC DEFINITIONS

The single scattering of light by a small-volume element dv consisting of randomly oriented, rotationally symmetric, independently scattering particles is completely described by the ensemble-averaged extinction, C_{ext}, and scattering, C_{sca}, cross sections per particle and the dimensionless

† Author to whom correspondence should be addressed.

$$p_{mn} = \sum_{n'=1}^{\infty} \sum_{m'=-n'}^{n'} \left(T^{11}_{mnm'n'} a_{m'n'} + T^{12} \ldots \right)$$

$$q_{mn} = \sum_{n'=1}^{\infty} \sum_{m'=-n'}^{n'} \left(T^{21}_{mnm'n'} a_{m'n'} + T \ldots \right)$$


Applicability of regular particle shapes in light scattering calculations for atmospheric ice particles

Andreas Macke and Michael I. Mishchenko

We ascertain the usefulness of simple ice particle geometries for modeling the intensity distribution of light scattering by atmospheric ice particles. To this end, similarities and differences in light scattering by axis-equivalent, regular and distorted hexagonal cylindric, ellipsoidal, and circular

erably larger than the wavelengths of the incoming solar radiation, especially in the visible spectral region. Therefore, the geometrical optics approximation offers a conceptually simple although time-consuming way to simulate single scattering by almost arbitrarily shaped scatterers.[1–5] Whereas these papers take more and more complex particle geometries such as bullet rosettes, dendrites, or polycrystals into account, in this paper we examine the possibility of representing the scattering proper-

On the other hand, the three (two) semiaxes of an ellipsoid (circular cylinder) allow for a variability of particle shapes that may cover to some extent the natural variability of atmospheric ice crystal habits.

Another motivation arises from uncertainties in our knowledge of real ice particle shapes. The study of observationally derived two-dimensional ice crystal shadow images[6] or replicas[7,8] clearly demonstrates that solid hexagonal columns or plates are a strong idealization of atmospheric ice crystals. However, statistically reliable shape information is difficult to extract from these data, partly because of the strong natural variability. Therefore it appears reasonable to ascertain the use of nonhexagonal but still simple geometries as substitutes for a polydispersion of complicated ice particle shapes.

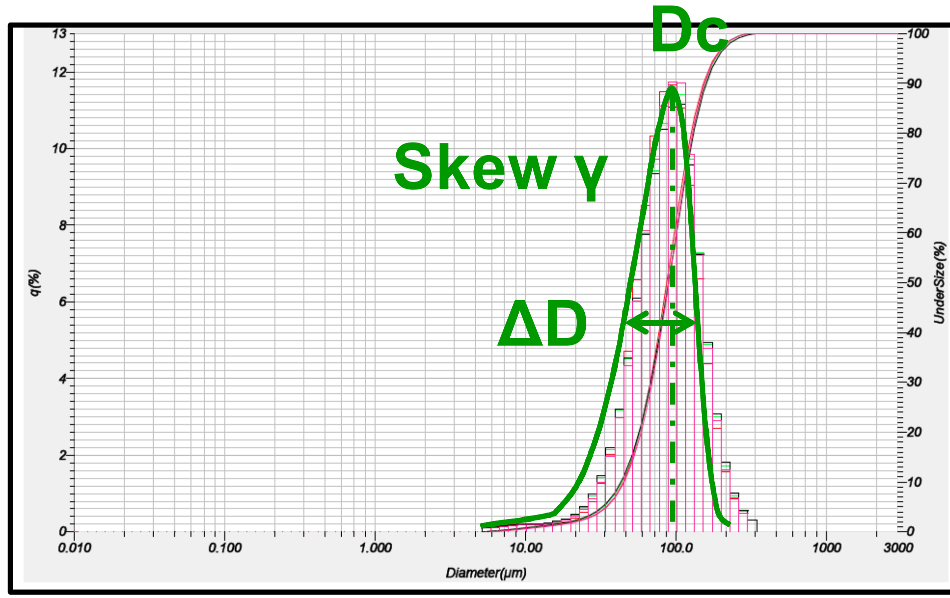
Because of the lack of sharp edges, ellipsoids do not provide strong halos that are characteristic of regular hexagonal particles. However, the absence of these features, as reported in a number of radiance measurements in or above cirrus clouds,[9,10] emphasizes the potential use of nonhexagonal par-

The authors are with the NASA Goddard Institute for Space Studies, 2880 Broadway, New York, New York 10025. A. Macke is also with the Department of Applied Physics, Columbia University, 2880 Broadway, New York, New York 10025. M. I. Mishchenko is also with the Institute of Terrestrial and Planetary Atmospheres, State University of New York at Stony Brook, Stony Brook, New York 11794.
Received 28 August 1995; revised manuscript received 29 January 1996.
0003-6935/96/214291-06$10.00/0

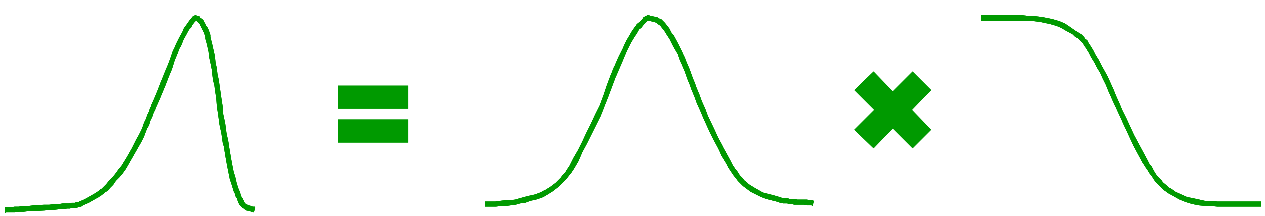
$\pi D/\lambda$ > 120 → **+ Diffraction**

Particle Size Distribution (PSD)

- Approximate with skewed *area* log-normal distribution
- Expression with 3 parameters: r_g, σ_g, γ



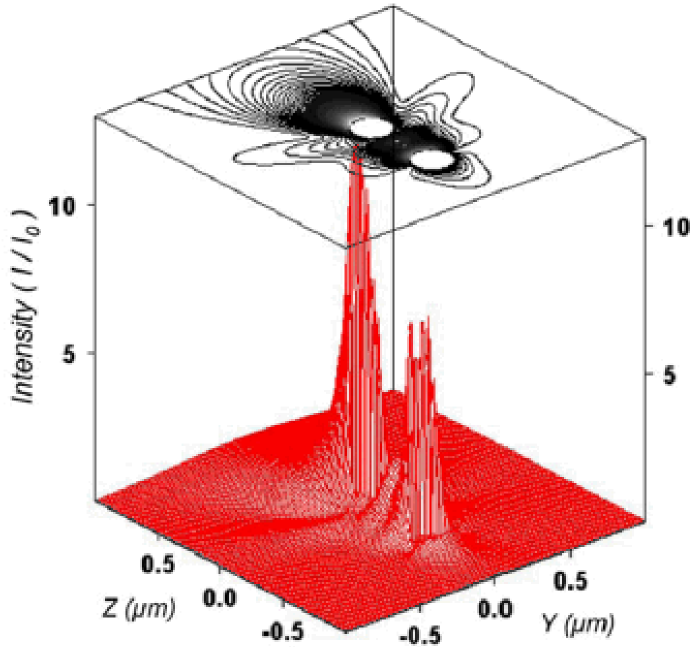
$$n(r) = \text{constant} \times r^{-3} \exp\left[-\frac{\left(\ln r - \ln r_g\right)^2}{2\ln^2 \sigma_g}\right] \times 2\left[1 + \text{erf}\left(\frac{\alpha\left(\ln r - \ln r_g\right)^2}{2\sqrt{2}\ln^2 \sigma_g}\right)\right]$$



“Patching” the Scattering Properties

- The radiative transfer equation (RTE) is strictly applicable only for sparse media (packing density < 1%)
- ***S(θ): Static structure factor (SSF)***
  - Acts as a multiplier to the scattering cross section and phase function
  - Analytical expression available for monodisperse spheres

$$p(\theta) = \frac{4\pi}{C_{\text{sca}}} \frac{dC_{\text{sca}}}{d\Omega} S(\theta)$$



J.-C. Auger and B. Stout, “Local field intensity in aggregates illuminated by diffuse light: T matrix approach,” Appl. Opt. **47**, 2897-2905 (2008).

“Patching” the Scattering Properties

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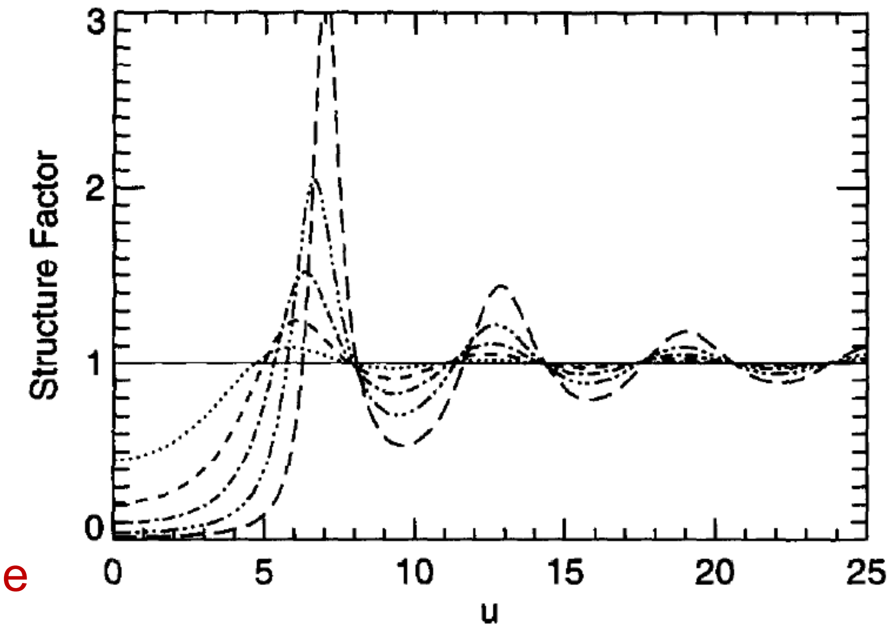
- ***S(θ): Static structure factor (SSF)***
  - Acts as a multiplier to the scattering cross section and phase function
  - Analytical expression available for monodisperse spheres
  - *$S(\theta) = F(f_{SSF}, u)$*
  - $u = \frac{8\pi r_0}{\lambda} \sin\left(\frac{\theta}{2}\right) = 4\left(\frac{\pi D}{\lambda}\right) \times \sin\left(\frac{\theta}{2}\right)$

Size parameter

Scattering angle

$$p(\theta) = \frac{4\pi}{C_{\text{sca}}} \frac{dC_{\text{sca}}}{d\Omega} S(\theta)$$



M. I. Mishchenko, “Asymmetry parameters of the phase function for densely packed scattering grains,” JQSRT **52**, 95-110 (1994).

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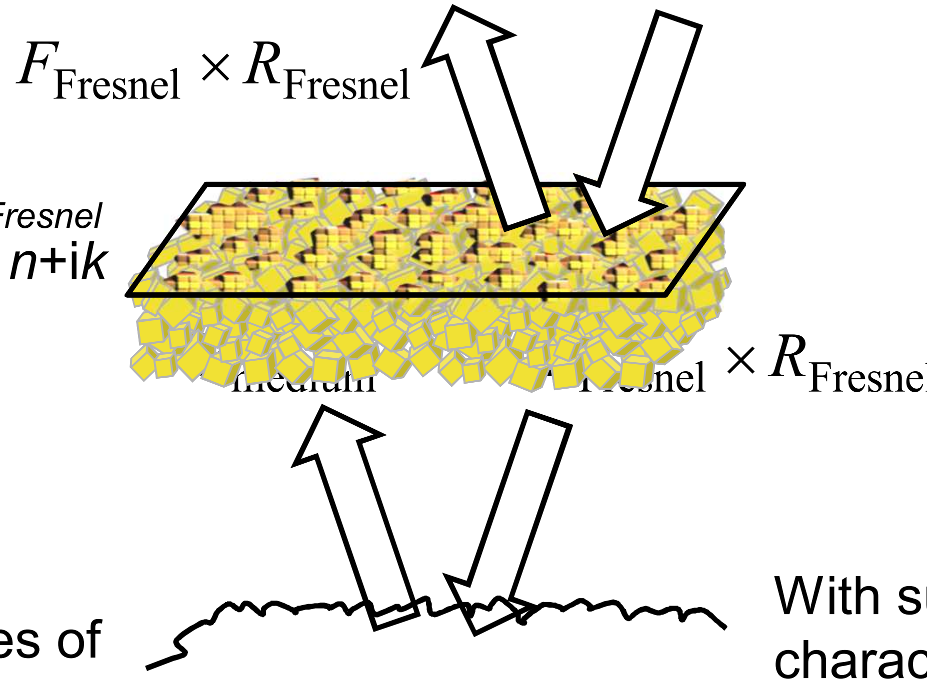
1st-Surface Reflectance and Surface Roughness

Fractional area $F_{Fresnel}$ of refractive index n+ik

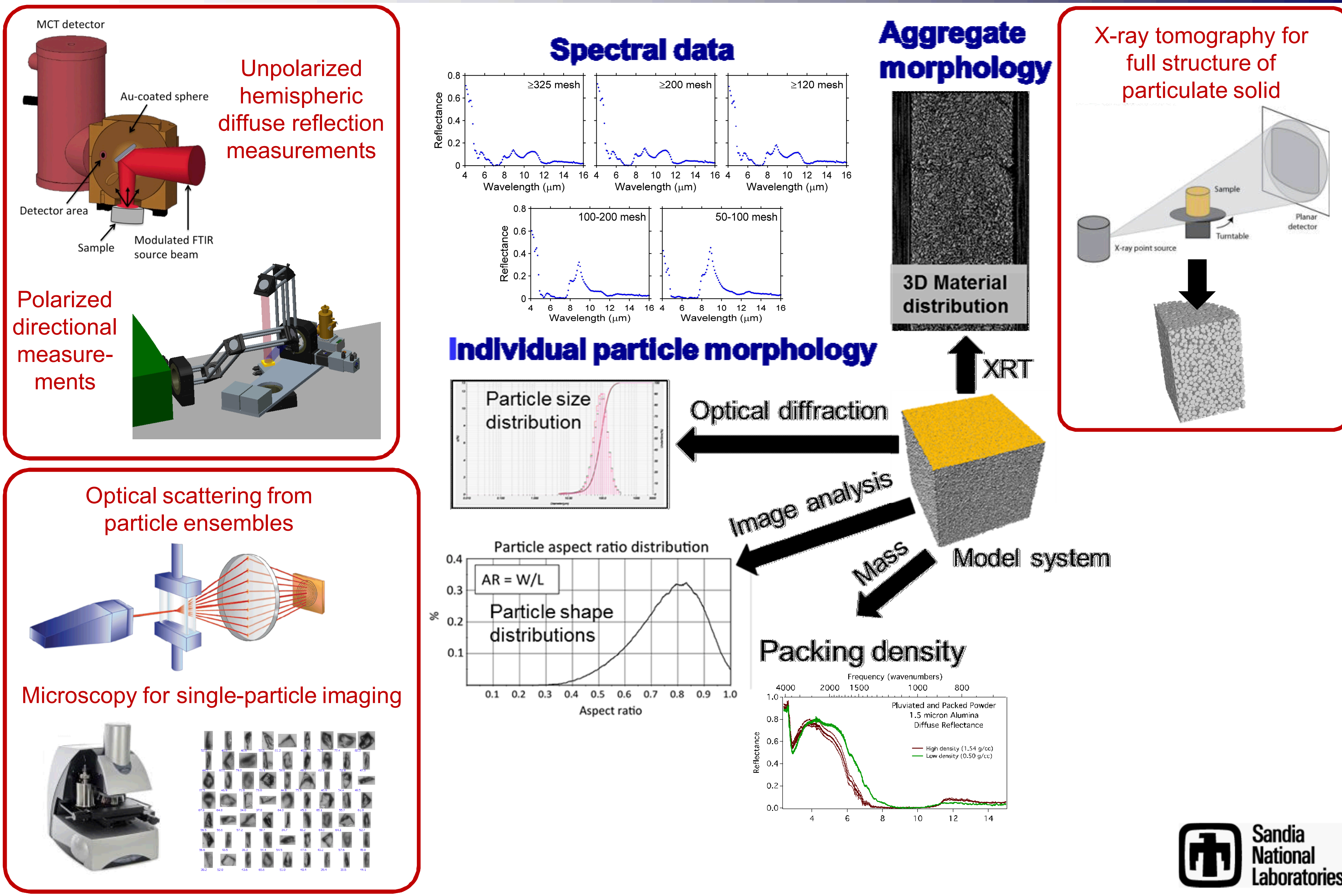


Medium of particles of refractive index n+ik

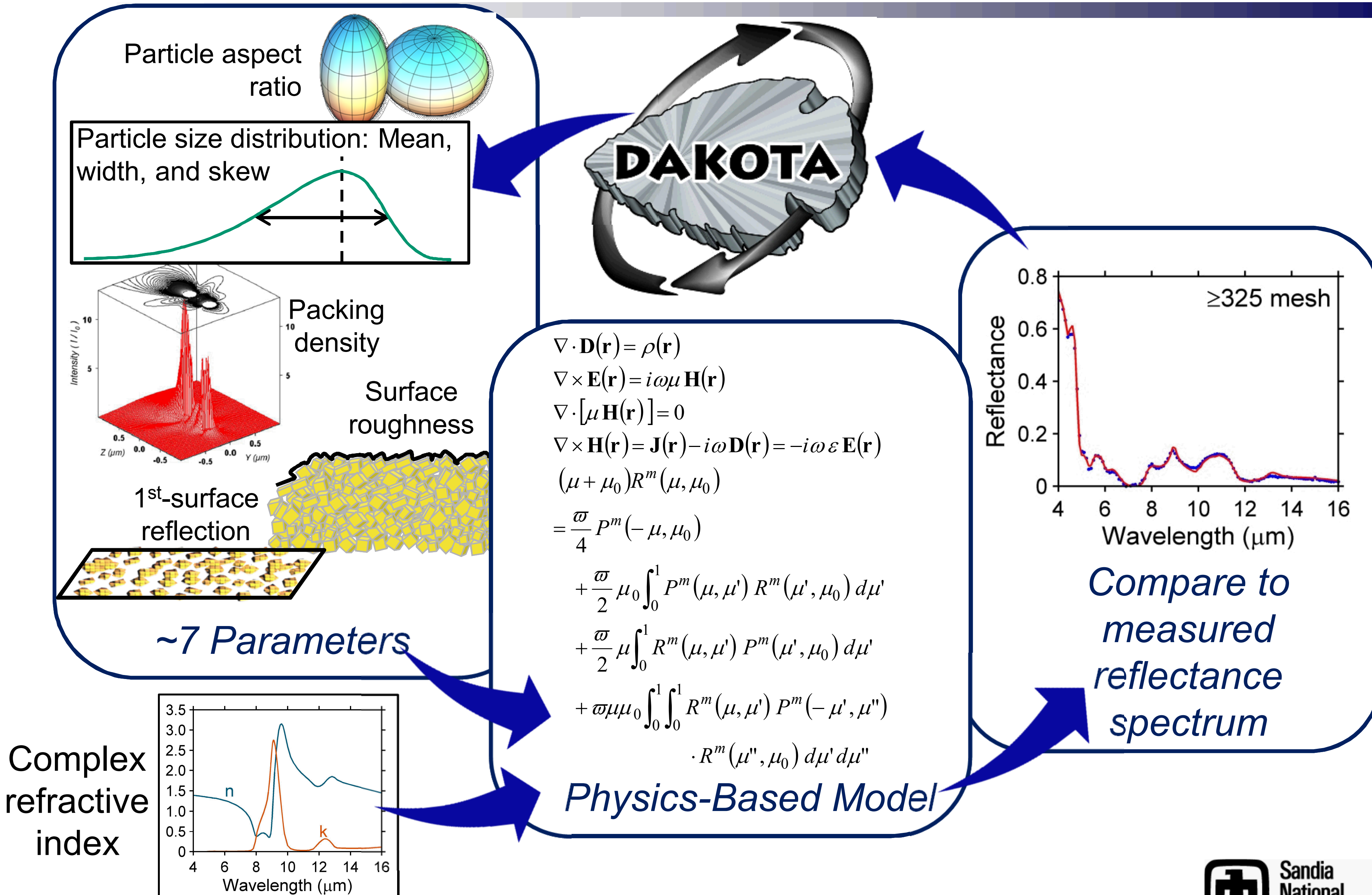
With surface roughness characterized by θ_{eff}

$$R = F_{\mathrm{Fresnel}} \times R_{\mathrm{Fresnel}} + \left(1 - F_{\mathrm{Fresnel}} \times R_{\mathrm{Fresnel}}\right) \times R_{\mathrm{medium}}$$

Full Characterization of Material Systems



Model Assessment



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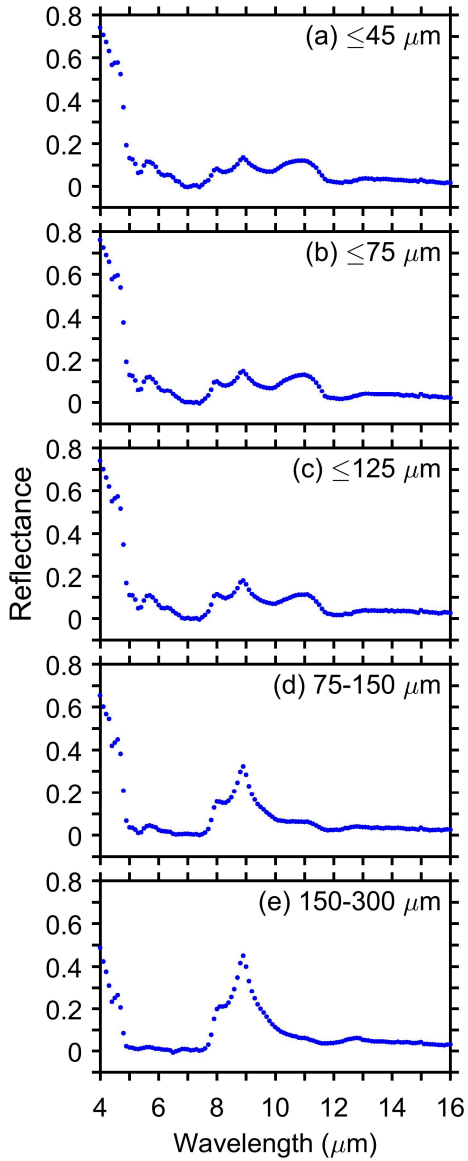
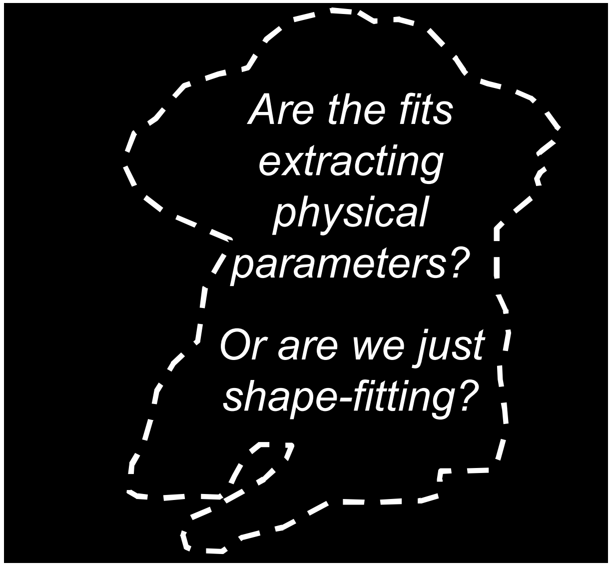
Measured and Modeled Reflectance Spectra of Silica Powders

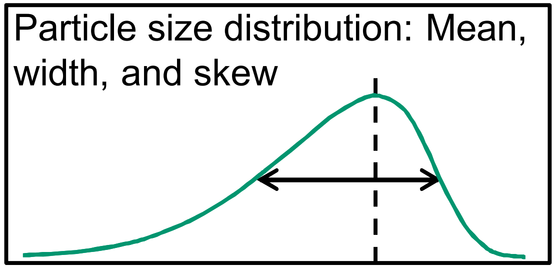


Nice agreement, but…

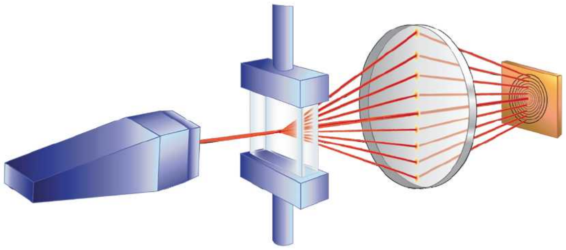
"Give me seven free parameters and I can fit an elephant"

– K. Lumme and A. Penttilä, JQSRT **112**, 1658-1670 (2011).

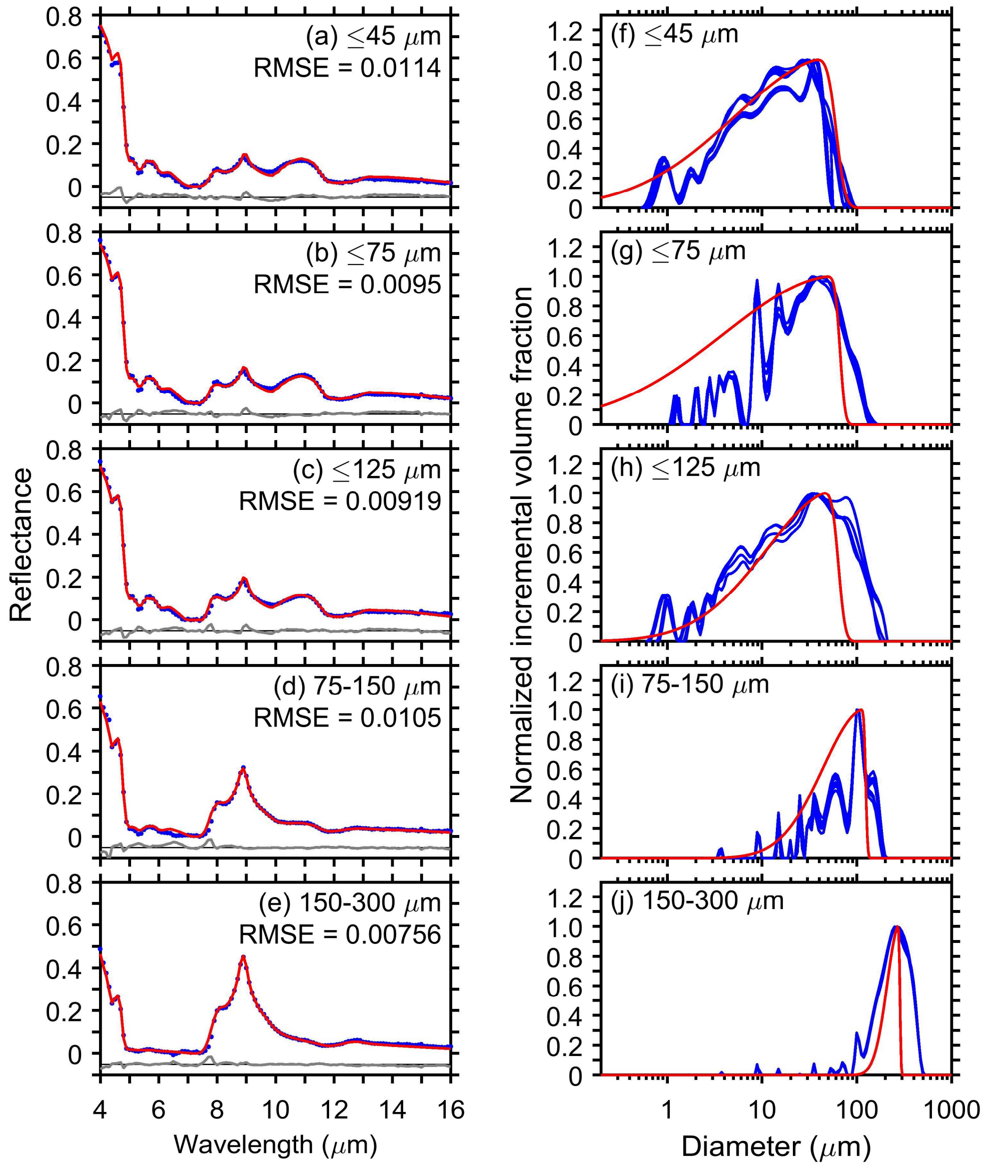




VS



Measured and Modeled Reflectance Spectra and PSDs of Silica Powders



- PSDs derived via optimizing reflectance model to measured spectra demonstrate general agreement with PSDs measured via laser diffraction
- Exact level of agreement confounded by broad distributions provided by manufacturer
- Repeat process on samples sieved in-house

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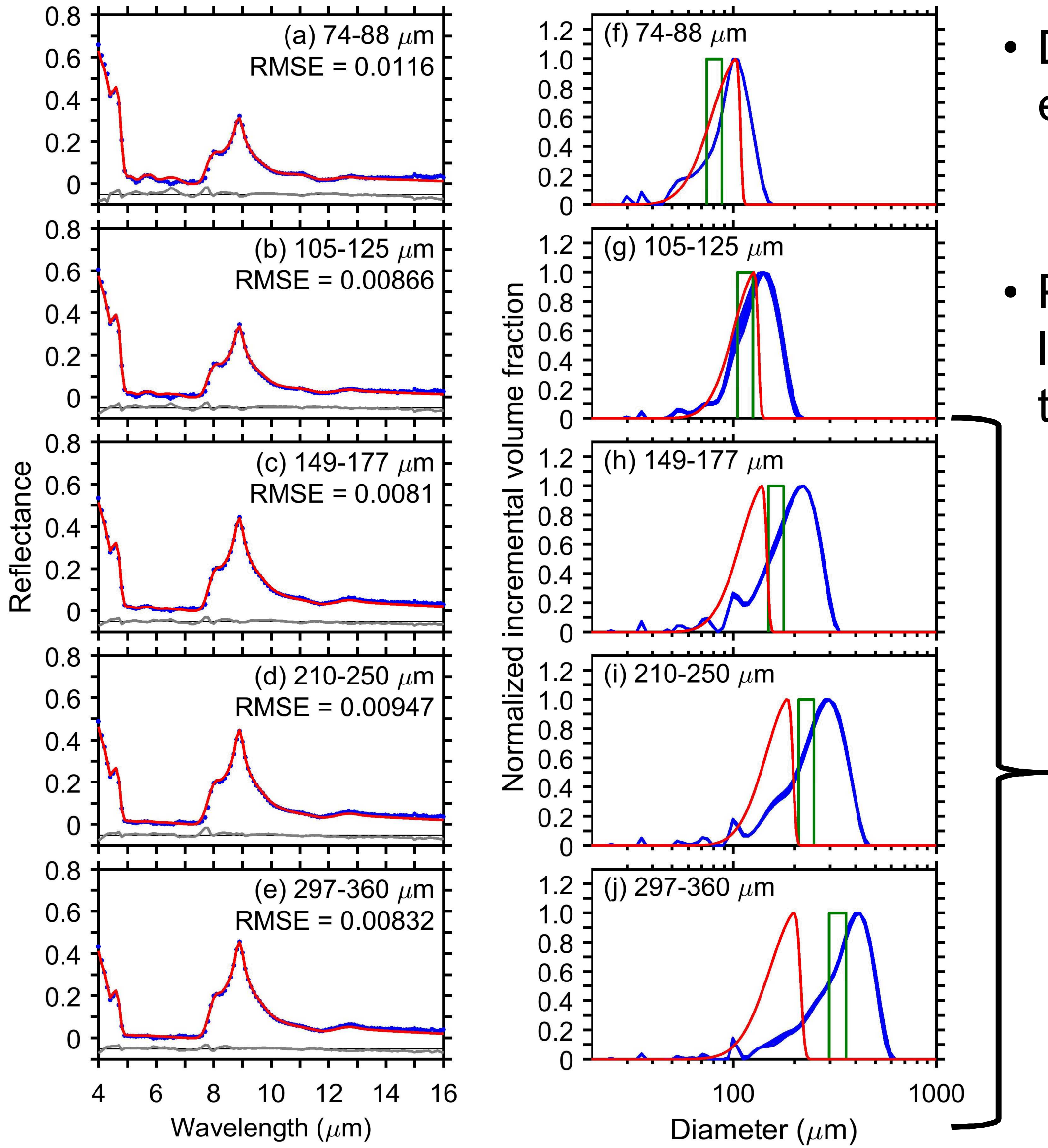
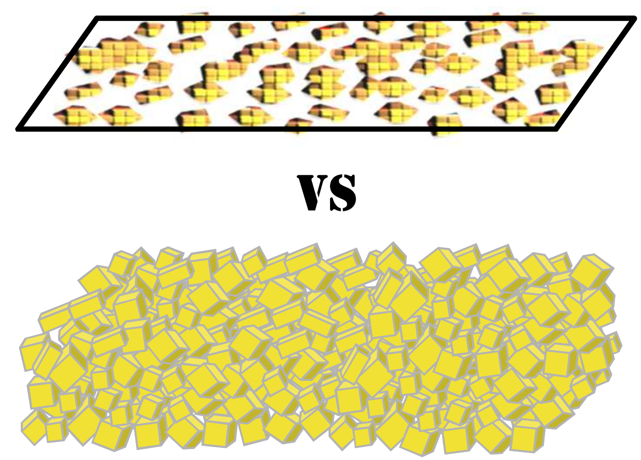
Measured and Modeled Reflectance Spectra and PSDs of Silica Powders



- Departure from measured PSDs is evident for larger particles
  - Model-derived PSDs are skewed to smaller particle sizes
- Potential cause: The reflectance of larger particles is spectrally similar to the $R_{Fresnel}$ of the bulk material
  - Optimization is likely attributing reflectance contribution of larger particles to 1st-surface reflectance term



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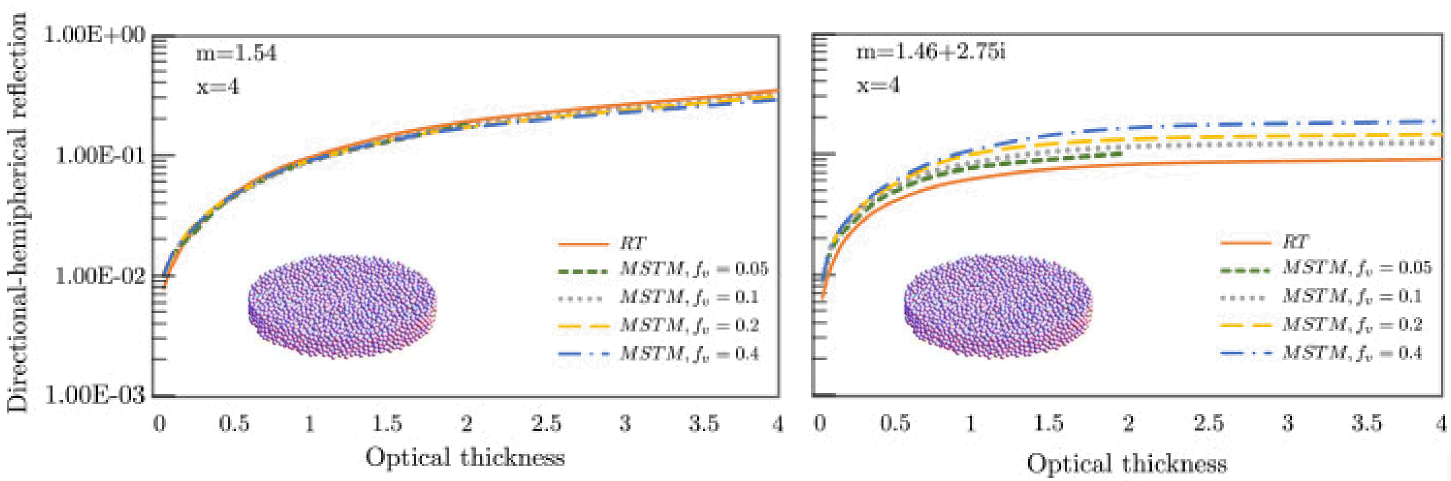
Summary and Next Steps

- Demonstrated physics-based model for the reflectance spectrum of an optically thick packed particulate medium
  - Requires as input both fundamental optical properties (*n*,*k*) and morphological parameters (PSD, packing density, surface roughness)
  - Optimized, the model spectra demonstrate agreement with measured spectra
  - Model-extracted PSDs agree with laser-diffraction measurements for smaller particles
- Further assessment(s) required
  - Model fully demonstrated (with rigorous PSD comparison) on only one material (silica)
  - Approximation for 1st-surface reflection requires further vetting, and new computational results could serve to inform this process

B. Ramezan pour and D. W. Mackowski, “Radiative transfer equation and direct simulation prediction of reflection and absorption by particle deposits,” JQSRT **189**, 361-368 (2017).



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Thanks! Questions?

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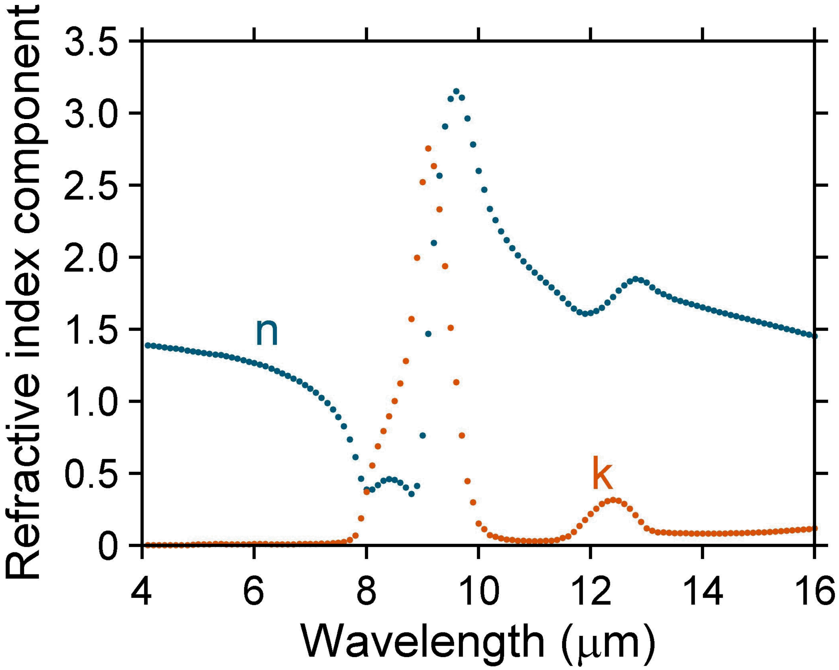
Extra Viewgraphs

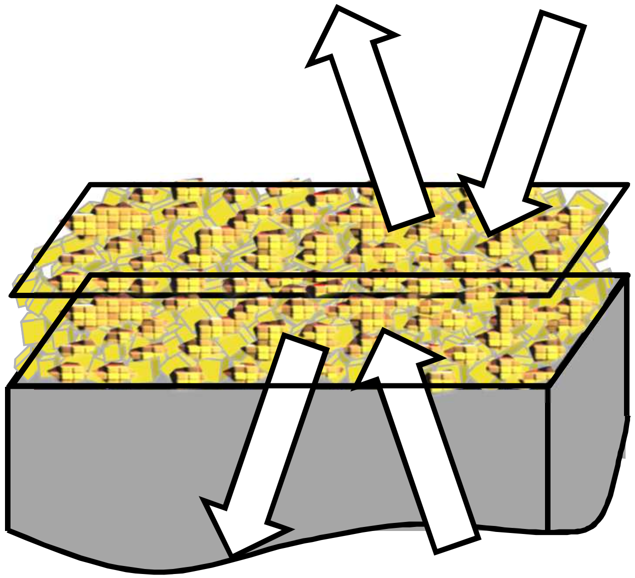
Optimization via "nl2sol" in DAKOTA

- 121 λs: 4-16 μm @ 0.1 μm resolution
- The 121 λ-dependent calculations are divided among 64 processors (4 nodes, 16 cores/node)
- ~10 min/spectrum, 100+ such calculations required for convergence



- The Jacobian (matrix of 1st-order partial derivatives) is numerically determined through forward difference calculations
- The Hessian (matrix of 2nd-order partial derivatives) is numerically approximated from special properties of the sum-of-squares

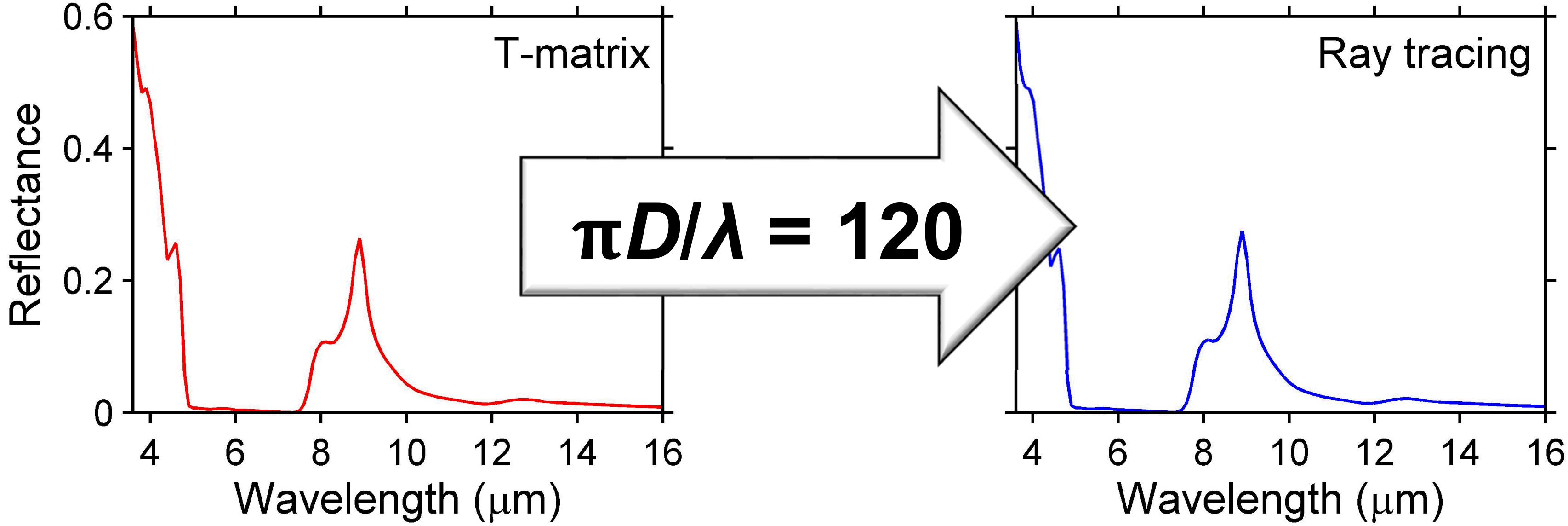
Transitioning to Optically Thin Materials



Should $R_{substrate}$ be modified to account for surface reflection from deposit?

$R_{substrate}$

T-matrix vs. Ray tracing



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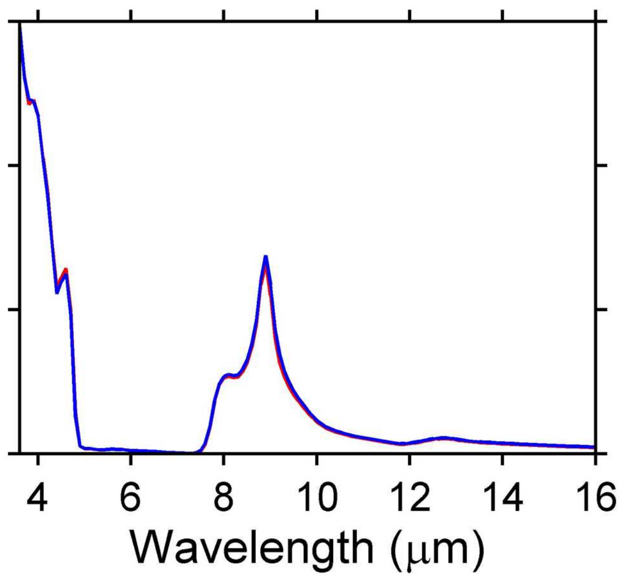
T-matrix vs. Ray tracing



Similar Approximations Made by Others

- J. L. Bandfield, P. O. Hayne, J.-P. Williams, B. T. Greenhagen, and D. A. Paige, “Lunar surface roughness derived from LRO Diviner Radiometer observations,” Icarus **248**, 357-372 (2015).
- P. Helfenstein and M. K. Shepard, “Submillimeter-scale topography of the lunar regolith,” Icarus **141**, 107-131 (1999).
- M. K. Shepard, R. A. Brackett, and R. E. Arvidson, “Self-affine (fractal) topography: Surface parameterization and radar scattering,” J. Geophys. Res. **100**, 11709-11718 (1995).
- B. Hapke, “Bidirectional reflectance spectroscopy. 3. Correction for Macroscopic Roughness,” Icarus **59**, 41-59 (1984).

But is this approximation sufficient?